$^{1)}$ At energies W not exceeding 107 eV, only slow electrons were emitted, and their energy distribution was in agreement with the Demkov-Komarov theory [5].

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## COEXISTENCE OF FERRO- AND ANTIFERROMAGNETIC ORDER IN INVAR IRON-NICKEL ALLOYS

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> A neutron-diffraction method was used to investigate the magnetic structure of the disordered alloy Fe<sub>0.63</sub>Ni<sub>0.37</sub>. It is shown for the first time that an antiferromagnetic order of the  $(\frac{1}{2}, \frac{1}{2}, 0)$ type is produced at liquid-helium temperature on top of the usual ferromagnetic order. The Neel point TN determined by measuring the temperature dependence of the peak intensity of the antiferromagnetic reflection was found to equal 15°K. The measurements results are interpreted within the framework of the noncollinear magnetic struc-

To explain the properties of invars, Kondorskii [1] advanced the hypothesis that in ferromagnetic Fe-Ni alloys with FCC crystal lattice the exchange integrals satisfy the relation

$$I_{11} > 0; I_{12} > 0; I_{22} < 0,$$
 (1)

where the subscripts 1 and 2 stand for the nickel and iron atoms, respectively. This hypothesis was confirmed experimentally in [2].

A canted-magnetic-structure model proposed in [3] for binary disordered alloys in which the exchange integrals satisfy the condition (1) has made it possible to explain the anomalous concentration dependences of the average magnetic moment per alloy atom in Fe-Ni alloys at 0°K [4], of the Curie temperature [5], and of the small-angle neutron scattering [6], and also the temperature dependences of the magnetization and of the coefficient of linear expansion [5].

This model suggests that ferromagnetic and antiferromagnetic long-range order can coexist in invar alloys at low temperatures (since  $I_{22}$  is appreciably smaller than  $I_{11}$  and  $I_{12}$ ), so that the mean values of the projection of the magnetic moment on the direction of the spontaneous magnetization form a ferromagnetic structure, while the projections of the magnetic moment on the perpendicular direction form an antiferromagnetic structure.

The presence of an antiferromagnetic transformation in Fe-Ni invar alloys is indirectly corroborated also by the data of [7].

To confirm the hypothesis experimentally, we have performed x-ray diffraction experiments on a disordered Fe-Ni alloy containing 63 at.% Fe in the temperature interval from 4.2 to 30°K, using a procedure described in [8].

The wavelength  $\lambda$  of the neutrons obtained by reflection from the lead crystal-monochromator was 1.07 Å. The admixtures of neutrons with  $\lambda/2$  and  $\lambda/3$  were 0.5 and 0.25%, respectively.

Figure 1b shows a reflection obtained from the sample at 4.2°K (this reflection is completely missing from the neutron diffraction pattern obtained at room temperature). To compare its relative intensity, Fig. 1c shows the (111) peak, to which nuclear and ferromagnetic scattering make contributions. As seen from Fig. 1a, which shows the temperature dependence of the peak intensity of the first reflection, the intensity decreases by more than one-half at 15°K, after which it falls off slowly with rising temperature.

In our opinion, the reflection shown in Fig. 1b is of antiferromagnetic origin. Favoring the antiferromagnetic nature of this reflection is the temperature dependence of its intensity, with a transition point near 15°K, and also the angular position corresponding to the largest distance between planes, which can be attributed to multiple increase of the magnetic unit cell in comparison with crystal-chemical one.

The best agreement with the experimental data is observed for a magnetic unit cell doubled in the x and y directions, as shown in Fig. 2.

The terms of the magnetic moments 1 and 2 in the z direction form a ferromagnetic order, and by virtue of their equivalence over all the sites Fig. 2a shows arbitrarily their magnitude in direction only at the origin. In accordance with [3, 4], the mean value (averaged over all the crystal-lattice sites) of the z-projection of the magnetic moment of the alloy at 0°K can be expressed in the form

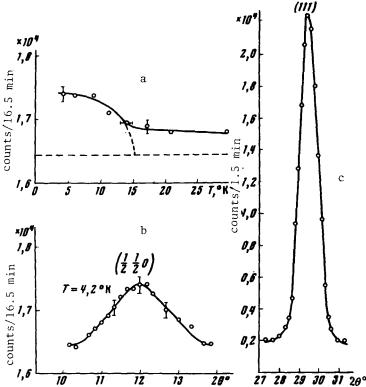


Fig. 1. a) Temperature dependence of the peak intensity of the reflection  $(\frac{1}{2}\frac{1}{2}0)$ . b) The reflection  $(\frac{1}{2}\frac{1}{2}0)$  at  $T = 4.2^{\circ}K$ . c) The reflection (111) at  $T = 4.2^{\circ}K$ .

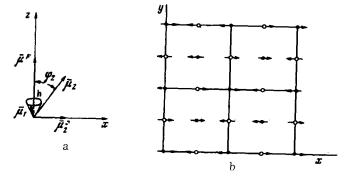


Fig. 2. a) Orientation of mean values of the magnetic moments of iron and nickel relative to the z direction in the site 000. b) Antiferromagnetic ordering of fourth kind for the perpendicular components of the magnetic moments of iron. The dark circles pertain to atoms at the level z=0, and the light ones to atoms at the level z=a/2.

$$\overline{\mu}^{\mathsf{F}} = \cos \phi_1 (1 - \epsilon) \mu_1 + \cos \phi_2 \epsilon \mu_2 = \cos \phi_1 \overline{\mu}_1 + \cos \phi_2 \overline{\mu}_2, \tag{2}$$

where c is the concentration of atoms 2 in the alloy, equal to the probability of substituting the lattice site of atoms 2;  $\mu_1$  = 0.6 $\mu_B$  and  $\mu_2$  = 2.8 $\mu_B$  are the magnetic moments of the nickel and iron atoms, respectively;  $\phi_1$  and  $\phi_2$  are the effective angles of deviation of the magnetic moments of the Ni and Fe atoms from the direction of spontaneous magnetization; the method of

Property	ũ <sup>F</sup> , μ <sub>B</sub>	$T_c / T_1$	μ̄A, μ <sub>B</sub>	$T_N/T_1$
Calc.	1,75	1,0	0,85	~ 0,05
Expt.	1,72 ± 0,05	0,95 ± 0,05	0,63 ± 0,20	0,024.±0,002

calculating these angles is given, for example, in [4].

As seen from the table, the experimental values of  $\tilde{\mu}^F$  and of the Curie temperature T<sub>c</sub> of the alloy (referred to the Currie temperature T1 of the nickel), determined from the data on the saturation magnetization, are in good agreement with the values calculated from formula (2) and from formula (14) of [5].

The perpendicular components of the magnetic moments of the iron atoms, as seen from Fig. 2b, form an antiferromagnetic order of the fourth kind in the FCC lattice.

The perpendicular components of the magnetic moments of the nickel atoms should apparently not form an antiferromagnetic order. Since any regular arrangement of these atoms relative to the corresponding projections of the iron moments greatly lowers the Neel temperature, by virtue of conditions (1), such an arrangement is not favored energywise.

The direction of the antiferromagnetic vector was chosen to lie along [100], inasmuch as according to [9] the subsequent resolved reflections (1/2 1/2 1) and (1/2 3/2 0) should in this case be much weaker than the reflection (1/2 1/2 0) in accord with the experimental data.

The antiferromagnetic component of the magnetic moment, determined from the intensity of the reflection (1/2 1/2 0), is in satisfactory agreement with the calculated value  $\bar{\mu}_{\Delta}^{A}$  = sin  $\phi_{2}$ c $\mu_{2}$ (see the table).

It is easy to write down an expression for the relative Neel temperature for the structure shown in Fig. 2b:

$$T_N/T_1 = -c \sin \phi_2 \frac{S_2(S_2+1)}{S_1(S_1+1)} \frac{2}{3} \frac{\gamma_{22}}{\gamma_{11}}$$
, (3)

where c sin  $\phi_2$  is the effective fraction of the atoms of sort 2, having a spin S = 3/2. S<sub>1</sub> = 1/2 is the spin of the nickel atoms; the factor 2/3 takes into account the fact that four out of the six magnetic moments in the second coordinate sphere are antiparallel and two are parallel to the moment located at the origin;  $\gamma_{22}/\gamma_{11}$  is the ratio of the molecular-field constants, which can be approximately assumed to equal -0.05 in accord with [5].

As seen from the table, the calculated value of  $T_{\rm N}/T_{\rm 1}$  agrees qualitatively with the experimentally observed one.

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